

Docket No.: M1071.1855  
(PATENT)

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

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In re Patent Application of:

Katsuhiro Horikawa et al.

Application No.: 10/624,537

Confirmation No.: 6662

Filed: July 23, 2003

Art Unit: 1734

For: MANUFACTURING METHOD FOR  
MONOLITHIC PIEZOELECTRIC PART,  
AND MONOLITHIC PIEZOELECTRIC  
PART

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Examiner: M. C. Mayes

**APPEAL BRIEF**

MS Appeal Brief - Patents  
Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Dear Sir:

As required under § 41.37(a), this brief is filed within two months of the Notice of Appeal filed in this case on December 11, 2006, and is in furtherance of said Notice of Appeal.

The fees required under § 41.20(b)(2) are dealt with in a separate paper.

This brief contains items under the following headings as required by 37 C.F.R. § 41.37 and M.P.E.P. § 1206:

- I. Real Party In Interest
- II. Related Appeals and Interferences
- III. Status of Claims
- IV. Status of Amendments
- V. Summary of Claimed Subject Matter
- VI. Grounds of Rejection to be Reviewed on Appeal
- VII. Argument
- VIII. Claims
- Appendix A Claims
- Appendix B Evidence
- Appendix C Related Proceedings

I. REAL PARTY IN INTEREST

The real party in interest for this appeal is:

MURATA MANUFACTURING CO., LTD.

II. RELATED APPEALS, INTERFERENCES, AND JUDICIAL PROCEEDINGS

There are no other appeals, interferences, or judicial proceedings which will directly affect or be directly affected by or have a bearing on the Board's decision in this appeal.

III. STATUS OF CLAIMS

A. Total Number of Claims in Application

There are 20 claims pending in application.

B. 4Current Status of Claims

1. Claims canceled: 2, 3, 10, and 17-20
2. Claims withdrawn from consideration but not canceled: 0
3. Claims pending: 1, 4-9, 11-16, and 21-27
4. Claims allowed: 0
5. Claims rejected: 1, 4-9, 11-16, and 21-27

C. Claims On Appeal

The claims on appeal are claims 1, 4-9, 11-16, and 21-27

IV. STATUS OF AMENDMENTS

Applicant did not amend the claims after Final Rejection.

V. SUMMARY OF CLAIMED SUBJECT MATTER

The presence of internal electrodes within a piezoelectric ceramic body has led to the deterioration of piezoelectric properties and also poor reliability as attempts have been made to reduce the size of monolithic piezoelectric parts by the devise of reducing the thickness of the layers of the ceramic used to make such parts and increasing the number of sheets. One property which deteriorated is the piezoelectric d constant, a reflection of the amount of mechanical strain produced by an applied electric field. However, a high piezoelectric d constant is required in certain monolithic piezoelectric parts such as monolithic piezoelectric actuators and audio emitters.

It is also recognized in the art that when the piezoelectric ceramic is a lead perovskite type material, the piezoelectric d constant conspicuously deteriorates when sintering takes place under low oxygen atmosphere conditions because that low oxygen concentration promotes the generation of oxygen pores. When the oxygen concentration is reduced to less than 1 percent by volume or when a soft piezoelectric ceramic material with a high piezoelectric d constant is used, the deterioration is so severe that it is difficult to even form the desired monolithic piezoelectric part.

The present invention is based, in part, on the discovery that a layered article having a plurality of piezoelectric ceramic layers and spaced internal electrode layers disposed in the part can be fabricated without deteriorating the piezoelectric d constant by co-sintering the ceramic layers and internal electrode layers in an atmosphere where the oxygen concentration is present in an positive amount up to about 5 volume percent, when the molar quantity of the A site of the perovskite crystal structure has been reduced by about 0.5 to 5 mole percent from stoichiometric, and when the average valence of the B site component is increased to greater than 4 but less than 4.1. The fact that the deterioration of the piezoelectric d constant can be combated by so doing is surprising and unexpected.

Independent claim 1 on appeal is a combination of original claims 1-3 and 10, and such original claims constitute one mapping of the present claim. Independent claim 22 is the same as current claim 1 except that instead of being drawn to making a ceramic actuator part, claim 22 specifies the part as a ceramic audio emitter part, basis for which can be found, *inter alia*, at page 1, line 8.

## VI. GROUNDS OF REJECTION TO BE REVIEWED ON APPEAL

There are 5 separate rejections in this appeal. They are:

Claims 1, 4, 5, 7-9, 11 and 14-16 are rejected under 35 U.S.C. § 103 over Horikawa (US 6,080,328) in view of Ponomarev (US 2004/0012000), Horikawa (US 6,383,408) and JP 2-122511.

Claims 6 and 21 are rejected under 35 U.S.C. § 103 over the same combination of references (Horikawa '328, Ponomarev, Horikawa '408, and JP '511) in further combination with Feltz (US 2002/098333).

Claims 12 and 13 are rejected under 35 U.S.C. § 103 over Horikawa '328, Ponomarev, Horikawa '408, and JP '511 in further combination with JP 2001-181035.

Claims 22 – 26 are rejected under 35 U.S.C. § 103 over Horikawa '328, Ponomarev, Horikawa '408, and JP '511 in further combination with Ogawa (US 6,280,650) or Takeshima (US 2001/0045792).

Claim 27 is rejected under 35 U.S.C. § 103 over Horikawa '328, Ponomarev, Horikawa '408, and JP '511 in further combination with JP '035.

## VII. ARGUMENT

The present invention relates to manufacture of piezoelectric actuators and other piezoelectric ceramic parts. As with other electronic materials, there has been a trend to smaller and smaller devices in recent years. That has given rise to problems and the current invention is directed to solving one of those problems.

More particularly, one method of reducing the size of monolithic piezoelectric parts is to increase the number of the layers of the ceramic which are formed into a laminate while reducing the thickness of the individual sheets at the same time, such that the total thickness is smaller. However, when there are internal electrodes within these piezoelectric ceramic bodies, the result has been the

deterioration of desired piezoelectric properties and reliability of the parts became poor. One property which deteriorated is the piezoelectric  $d$  constant, a value which reflects the amount of mechanical strain produced by an applied electric field. This is quite undesirable because a high piezoelectric  $d$  constant is required in certain monolithic piezoelectric parts such as monolithic piezoelectric actuators and audio emitters.

It is also recognized in the art that when the piezoelectric ceramic is made of a lead perovskite type material, that is a ceramic having the general formula  $ABO_3$  (A and B represent sites of the crystal structure), the piezoelectric  $d$  constant conspicuously deteriorated when sintering of the perovskite took place in an atmosphere containing a low oxygen concentration because the low oxygen concentration promoted the generation of pores. Indeed, when the oxygen concentration is reduced to less than 1 percent by volume or when a soft piezoelectric ceramic material with a high piezoelectric  $d$  constant is used, the deterioration has become so severe that it is difficult to even form the piezoelectric ceramic into the desired monolithic piezoelectric part.

The present invention is based, in part, on the discovery that a layered article having a plurality of piezoelectric ceramic layers and spaced internal electrode layers disposed in the part can be fabricated without deteriorating the piezoelectric  $d$  constant by co-sintering the ceramic layers and internal electrode layers in an atmosphere where the oxygen concentration during sintering is low and does not exceed about 5 volume percent, if the molar quantity of the A site of the perovskite has been reduced by about 0.5 to 5 mole percent from the stoichiometric amount, and the average valence of the B site component is increased to greater than 4 but less than 4.1. The fact that the

deterioration of the piezoelectric d constant can be combated is surprising and unexpected.

A. Claims 1, 4, 5, 7-9, 11 and 14-16 are patentable under 35 U.S.C. § 103 over Horikawa '328 in view of Ponomarev, Horikawa '408 and JP '511

Horikawa '328 teaches making a piezoelectric ceramic element by forming a laminate of layers of a chromium-niobium-containing PZT (lead zirconate titanate) ceramic with internal electrodes disposed between adjacent layers sheets, followed by firing the laminate. The rejection relies on this reference to show co-sintering of a structure having laminated green (*i.e.*, unsintered) piezoelectric ceramic and internal electrode layers. It is acknowledged that this reference is deficient in that there is no disclosure of either altering the stoichiometry of the PZT or of sintering in an atmosphere of reduced oxygen content.

The Ponomarev reference has been cited solely to teach that low loss hard piezoelectric ceramic materials are desired in multi-layer ceramic transformers.

Horikawa '408 relates to low loss piezoelectric ceramics for high frequency filters and isolators. The ceramic is a manganese and niobium-containing PZT of the particular composition set forth at column 2, lines 50 to 55. This ceramic is used to prepare a ceramic molded body which is then sintered and thereafter processed to apply electrodes to the sintered body. There is no teaching or suggestion in this reference of cofiring a green (unsintered) piezoelectric ceramic body which has internal electrodes under any conditions. In addition, there is no teaching or suggestion that the ceramic should be fired in a reduced oxygen atmosphere for any reason. Accordingly, the ceramic of Horikawa '408 does not have to face the problem of deterioration of the piezoelectric d constant.

The combination of Horikawa '328, Ponomarev, and Horikawa '408 fails to teach or suggest that piezoelectric parts can be fabricated without deteriorating the piezoelectric  $d$  constant, a surprising and unexpected result, by co-sintering ceramic layers and internal electrode layers in an atmosphere where the oxygen concentration is a positive amount of up to about 5 volume percent, if the molar quantity of the A site of the perovskite has been reduced by about 0.5 to 5 mole percent from the stoichiometric amount, and the average valence of the B site component is increased to greater than 4 but less than 4.1. The combination does not teach or suggest employing this combination of features for any reason. The additional reference, JP '511<sup>1</sup> does not obviate this deficiency.

JP '511 concerns a ceramic capacitor made by sintering in a low oxygen atmosphere. The ceramics in this reference are either 97wt% TiO<sub>2</sub> – 2wt% CuO – 1wt% Zr (page 3) or 95wt% TiO<sub>2</sub> – 4wt% CuO – 1wt% Zr (page 4). These are dielectric ceramics, not piezoelectric ceramics. They do not contain lead nor are they perovskites nor are they PZT type ceramics nor are they piezoelectric ceramics. However, the other references proposed in this combination rejection can be broadly characterized as concerning a PZT system piezoelectric ceramic body. The crystal structure of a titanium dioxide type dielectric (as in JP '511) is very different from that of a perovskite piezoelectric (as in the other references). That means that there must be some reason or motivation to extract some part of the teachings of JP '511 relating to one type of crystal structure, and utilize it in the PZT piezoelectric ceramics of the other references, which have a different crystal structure, without using hindsight or the appealed claims as a template, but no reason or motivation is apparent from the references nor has any been proposed by the Examiner. The Office Actions simply

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<sup>1</sup> The Japanese Patent issued on the JP '511 application, namely Japanese Patent No. 2,676,620, is discussed on pages 2-3 of the present specification.



states it would be obvious to do modification “to improve reliability and reduce cost while retaining needed characteristics”. But the Office Actions never explain why one skilled in the art would expect measures taken to “improve reliability and reduce cost while retaining needed characteristics” of a dielectric, non-PZT system will “improve reliability and reduce cost while retaining needed characteristics” in a PZT piezoelectric system. The proffered motivation seems to assume without any factual basis that the “needed characteristics” are the same. Without such explanation, the justification advanced is simply a retrospective conclusion that an advantage can be achieved.

It is therefore clear that the reliance on JP ‘511 in this rejection can only be based on hindsight as well as using the appealed claims as a template. Neither is permissible.

Beyond the foregoing, the rejection seeks to ignore other art of record in this case even though the law requires such art be given consideration. In particular, Randall (US 2002/0079622) concerns co-firing a multilayered piezoelectric ceramic material with base metal electrodes, as done here, and acknowledges that there are significant problems attendant to such co-firing, including, *inter alia*, firing without oxidation of the electrodes and also without reduction of the PZT or other ceramic material. See e.g. paragraph [0007]. Watanabe (JP 11-163433) states that “it is important to reduce diffusion of Ag from an internal electrode” in a PZT type piezoelectric [0023] (whose thickness is not being reduced) and it is “desirable to make the oxygen density in a furnace at the time of sintering into 1-10%, for setting the content of Ag in electrostrictive ceramics to 10 ppm or more [but] less than 50 ppm” [0028], as a lower oxygen concentration gives rise to problems [0029]. In that connection, [0042] states that delamination occurred when the oxygen concentration was 0%. The prior art discussed on page 2 of the present application also teaches the skilled person that

piezoelectric transformers made with hard type ceramic and silver electrodes achieved good properties as a result of increasing the A site perovskite component content (the opposite of which is done in the invention). It also teaches the artisan that sintering a lead perovskite piezoelectric ceramic material in a reduced oxygen atmosphere to suppress silver dispersion in the ceramic led to significant deterioration in the piezoelectric d constant by generating oxygen pores to an extent which could even render the sintered ceramic unusable for its intended purpose. This fact is further illustrated in the comparative examples of Table 3 in the present application.

The record in this case thus clearly establishes that those skilled in the art would expect a lead perovskite piezoelectric ceramic material with internal electrodes to have a deteriorated piezoelectric d constant when sintered in a reduced oxygen atmosphere. The fact that the deterioration can be combated when making an actuator or emitter by employing a ceramic whose A and B site stoichiometric amounts have been altered as claimed is not taught or suggested and is, in light of the expectations of the art, surprising and unexpected.

This rejection should be reversed.

B. Claims 6 and 21 are patentable under 35 U.S.C. § 103 over the same combination further combined with Feltz

The combination of Horikawa '328, Ponomarev, Horikawa '408 and JP '511 has been discussed in the preceding section A. The additional Feltz reference has been cited only to teach that it is possible to do a partial substitution of the ions in the B position of the perovskite but it is not asserted to overcome, nor in fact does it overcome, the deficiencies in the prior combinations of references discussed above.

Accordingly, the further combination of these references with Feltz cannot render these claims obvious.

C. Claims 12 and 13 are patentable under 35 U.S.C. § 103 over the claim 11 references in combination with JP '035.

The combination of Horikawa '328, Ponomarev, Horikawa '408 and JP '511 has been discussed above. The additional Japanese reference has been cited only with regard to the internal electrodes and not to overcome the basic deficiencies in the base combination of references. It does not do so and reversal of this rejection is respectfully solicited.

D. Claims 22 – 26 are patentable under 35 U.S.C. § 103 over the Claim 1 applied references in combination with Ogawa or Takeshima.

The combination of Horikawa '328, Ponomarev, Horikawa '408 and JP '511 has been discussed above. These additional two references show that a buzzer could be a type of piezoelectric device. Such a teaching does not cure the basic deficiencies in the base combination of references, and cannot serve to render these claims obvious.

E. Claim 27 is patentable under 35 U.S.C. § 103 over Horikawa '328, Ponomarev, Horikawa '408, and JP '511 in further combination with JP '035.

The combination of Horikawa '328, Ponomarev, Horikawa '408 and JP '511 has been discussed above. The additional Japanese reference, JP '035 has been cited only with regard to the internal electrodes and not to overcome the basic deficiencies in the base combination of references. It does not do so and reversal of this rejection is respectfully solicited.

F. Conclusion

None of the rejections advanced in this appeal is viable and therefore, they should be reversed.

VIII. CLAIMS

A copy of the claims involved in the present appeal is attached hereto as Appendix A.

IX. EVIDENCE

An Appendix B is attached to indicate there is no such evidence.

X. RELATED PROCEEDINGS

An Appendix C is attached to indicate there are no such related proceedings.

Dated: February 12, 2007

Respectfully submitted,

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## APPENDIX A

### **Claims Involved in the Appeal of Application Serial No. 10/624,537**

1. A method for manufacturing a monolithic piezoelectric ceramic actuator part which has a plurality of piezoelectric ceramic layers and spaced internal electrode layers disposed in said piezoelectric ceramic actuator, wherein said piezoelectric ceramic making up said piezoelectric ceramic layers is formed of a perovskite compound oxide expressed by the general formula of  $ABO_3$ , and comprises at least Pb for the A site component and comprises Ti or Ti and Zr for the B site component and said internal electrode layers contain Ag as a primary component; said method comprising:

providing a piezoelectric ceramic powdered raw material wherein the molar quantity of said A site component is reduced by about 0.5 mol% to 5.0 mol% from that of a stoichiometric composition and the average valence of said B site component of the ceramic raw material is greater than that of the stoichiometric composition and is greater than 4.000 and less than 4.100;

fabricating a layered article with said piezoelectric ceramic powdered raw material; and

sintering said layered article within an atmosphere wherein the oxygen concentration is about 5% by volume or less but more than 0% by volume.

4. A method for manufacturing a monolithic piezoelectric actuator part according to Claim 1, wherein the molar quantity of Pb included in said A site component has been reduced by about 0.5 mol% to 5.0 mol% from that of the stoichiometric composition.

5. A method for manufacturing a monolithic piezoelectric actuator part according to Claim 4, wherein said B site component further comprises Nb.

6. A method for manufacturing a monolithic piezoelectric actuator part according to Claim 4, wherein said B site component further comprises Nb and Ni.

7. A method for manufacturing a monolithic piezoelectric actuator part according to Claim 4, wherein said B site component further comprises at least one of Nb, Sb, Ta and W.

8. A method for manufacturing a monolithic piezoelectric actuator part according to Claim 7, wherein said B site component further comprises at least one of Ni, Cr, Co and Mg.

9. A method for manufacturing a monolithic piezoelectric actuator part according to Claim 8, wherein said layered article fabrication comprises a ceramic green sheet fabrication forming said piezoelectric ceramic powdered raw material into sheet form so as to fabricate a plurality of ceramic green sheets, forming an electrode pattern on at least two of said ceramic green sheets with an electroconductive paste for internal electrodes which contains Ag as a primary component, and layering a plurality of ceramic green sheets upon which said electrode patterns have been formed so as to form a layered article.

11. A method for manufacturing a monolithic piezoelectric actuator part according to Claim 9, wherein said electroconductive paste contains Ag and Pd in a ratio of at least 70/30.

12. A method for manufacturing a monolithic piezoelectric actuator part according to Claim 11, wherein said electroconductive paste contains Ag and Pd in a ratio of at least 80/20 and the thickness of the ceramic layers is such that their thickness after sintering is about 64 $\mu$ m or less.

13. A method for manufacturing a monolithic piezoelectric actuator part according to Claim 12, wherein said electroconductive paste contains Ag and Pd in a ratio of at least 85/15 and the thickness of the ceramic layers is such that their thickness after sintering is about 40 $\mu$ m or less.

14. A method for manufacturing a monolithic piezoelectric part according to Claim 1, wherein said B site component comprises Ti and Zr.

15. A method for manufacturing a monolithic piezoelectric actuator part according to Claim 1, wherein said B site component further comprises Nb.

16. A method for manufacturing a monolithic piezoelectric actuator part according to Claim 1, wherein said layered article fabrication comprises a ceramic green sheet fabrication forming said piezoelectric ceramic powdered raw material into sheet form so as to fabricate a plurality of ceramic green sheets, forming an electrode pattern on at least two of said ceramic green sheets with an electroconductive paste for internal electrodes which contains Ag as a primary component, and layering a plurality of ceramic green sheets upon which said electrode patterns have been formed so as to form a layered article.

21. A method for manufacturing a monolithic piezoelectric actuator part according to Claim 1, wherein said B site component further comprises Nb and Ni.

22. A method for manufacturing a monolithic piezoelectric ceramic audio emitter part which has a plurality of piezoelectric ceramic layers and spaced internal electrode layers disposed in said piezoelectric ceramic audio emitter part, wherein said piezoelectric ceramic making up said piezoelectric ceramic layers is formed of a perovskite compound oxide expressed by the general formula of  $ABO_3$ , and comprises at least Pb for the A site component and comprises Ti or Ti and Zr for the B site

component and said internal electrode layers contain Ag as a primary component; said method comprising:

providing a piezoelectric ceramic powdered raw material wherein the molar quantity of said A site component is reduced by about 0.5 mol% to 5.0 mol% from that of a stoichiometric composition and the average valence of said B site component of the ceramic raw material is greater than that of the stoichiometric composition and is greater than 4.000 and less than 4.100;

fabricating a layered article with said piezoelectric ceramic powdered raw material; and

sintering said layered article within an atmosphere wherein the oxygen concentration is about 5% by volume or less but more than 0% by volume.

23. A method for manufacturing a monolithic piezoelectric ceramic audio emitter part according to Claim 22, wherein the molar quantity of Pb included in said A site component has been reduced by about 0.5 mol% to 5.0 mol% from that of the stoichiometric composition.

24. A method for manufacturing a monolithic piezoelectric ceramic audio emitter part according to Claim 23, wherein said B site component further comprises Nb.

25. A method for manufacturing a monolithic piezoelectric ceramic audio emitter part according to Claim 24, wherein said layered article fabrication comprises a ceramic green sheet fabrication forming said piezoelectric ceramic powdered raw material into sheet form so as to fabricate a plurality of ceramic green sheets, forming an electrode pattern on at least two of said ceramic green sheets with an electroconductive paste for internal electrodes which contains Ag as a primary



component, and layering a plurality of ceramic green sheets upon which said electrode patterns have been formed so as to form a layered article.

26. A method for manufacturing a monolithic piezoelectric ceramic audio emitter part according to Claim 25, wherein said electroconductive paste contains Ag and Pd in a ratio of at least 70/30.

27. A method for manufacturing a monolithic piezoelectric ceramic audio emitter part according to Claim 22, wherein said electroconductive paste contains Ag and Pd in a ratio of at least 80/20 and the thickness of the ceramic layers is such that their thickness after sintering is about 64μm or less.

**APPENDIX B**

No evidence pursuant to §§ 1.130, 1.131, or 1.132 or entered by or relied upon by the examiner is being submitted.

**APPENDIX C**

No related proceedings are referenced in II. above, hence copies of decisions in related proceedings are not provided.